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EMISSION FACTORS FOR SEVERAL TOXIC AIR POLLUTANTS
FROM FLUIDIZED-BED COMBUSTION OF COAL

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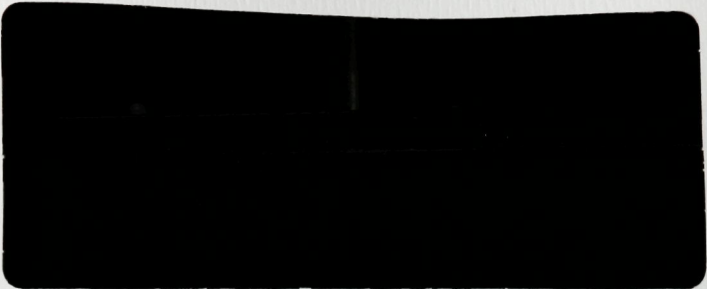


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This informal report presents preliminary results of ongoing work or work that is more limited in scope and depth than that described in formal reports issued by the Energy and Environmental Systems Division.

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ABSTRACT

1 INTRODUCTION

2 EMISSION FACTORS

ANL/EES-TM-327

2.1 Conventional Pollutants

2.2 Trace Elements

3 SUMMARY

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REFERENCES

BIBLIOGRAPHY

APPENDIX A: Parameter Correlations

APPENDIX B: Calculations and

by

Albert E. Smith

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Physical Environmental Sciences Section

1 Approximate Trace Element Emissions

2 Approximate Trace Element Emissions

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CONTENTS

ABSTRACT	1
1 INTRODUCTION	1
2 EMISSION FACTORS	3
2.1 Conventional Pollutants	3
2.2 Trace Elements	4
3 SUMMARY	8
REFERENCES	9
BIBLIOGRAPHY	10
APPENDIX A: Persons Contacted	11
APPENDIX B: Calculations and Assumptions	12

TABLES

1 Approximate Trace Element Emission Rates for AFBC	5
2 Approximate Trace Element Emission Rates for PFBC	7

EMISSION FACTORS FOR SEVERAL TOXIC AIR POLLUTANTS FROM FLUIDIZED-BED COMBUSTION OF COAL

by

Albert E. Smith

ABSTRACT

Clean coal technologies such as fluidized-bed combustion have the potential to emit the same trace elements as conventional combustors. Since the U.S. Environmental Protection Agency (EPA) is likely to promulgate National Emission Standards for Hazardous Air Pollutants for several trace elements, the feasibility of using fluidized-bed combustors to reduce sulfur dioxide emissions may depend in part on the relative amounts of trace elements emitted by fluidized-bed and conventional combustors. Emissions of trace elements from both atmospheric and pressurized fluidized-bed combustors were compared with those from conventional combustors by developing fluidized-bed emission factors from information available in the literature and comparing them with the emission factors for conventional combustors recommended in a literature search conducted for EPA. The comparisons are based on the mass of emission per unit of heat input for antimony, arsenic, beryllium, cadmium, chromium, cobalt, copper, lead, manganese, mercury, nickel, vanadium, and zinc. When inaccuracies in the data were taken into account, the trace element emissions from atmospheric fluidized-bed combustion seem to be somewhat higher than those from a conventional utility boiler burning pulverized coal and somewhat lower than those from pressurized fluidized-bed combustion.

1 INTRODUCTION

Clean coal technologies (e.g., gasification, liquefaction, and fluidized-bed combustion [FBC]) have received considerable attention as alternatives to conventional combustion of coal. Each clean coal technology has the potential to emit the same contaminants as conventional combustion. Under Sec. 112 of the Clean Air Act, the U.S. Environmental Protection Agency (EPA) is promulgating National Emission Standards for Hazardous Air Pollutants (NESHAPs) and is determining whether to propose NESHAPs for various trace elements. Since NESHAPs apply to all sources of a pollutant, both clean coal technologies and conventional combustors might be affected by NESHAPs that limit emissions of trace elements like cadmium, chromium, and nickel, all of which are found in coal. Knowing whether clean coal technologies have lower emissions of trace elements than conventional combustors would help in assessing possible barriers to the use of clean coal technologies. This study was undertaken to compare the trace element emissions from clean coal technologies with those from conventional combustion of coal.

On the basis of a preliminary literature search, the effort was limited to atmospheric and pressurized fluidized-bed combustion (AFBC and PFBC, respectively), there being an apparent paucity of easily usable information for the other clean coal technologies. The bibliography lists the literature searched for, but not cited in, this report.

A multipollutant risk assessment for conventional combustion sources is being conducted by EPA. By design, this study emphasizes the heavy metals that are being considered for inclusion in the EPA study (Mead et al., 1986). The results are presented as element-specific emission factors for ease of comparison with the emission factors being developed to support EPA's risk assessment.

1 INTRODUCTION

Clean coal technologies (e.g., gasification, fluidized-bed, and fluidized-bed combustion (FBC)) have received considerable attention as alternatives to conventional combustion of coal. Each clean coal technology has the potential to emit the same pollutants as conventional combustion. Under Section 112 of the Clean Air Act, the U.S. Environmental Protection Agency (EPA) is promulgating National Emission Standards for Hazardous Air Pollutants (NESHAPs) and is determining whether to promulgate NESHAPs for various trace elements. Since NESHAPs apply to all sources of a pollutant, both clean coal technologies and conventional combustion might be regulated by NESHAPs for these elements of trace elements like cadmium, chromium, and nickel, all of which are listed in Table 1. Knowing whether clean coal technologies have lower emissions of these elements than conventional combustion would help in evaluating whether the use of clean coal technologies. This study was undertaken to generate the trace element emissions from clean coal technologies with those from conventional combustion of coal.

2 EMISSION FACTORS

Conversations with staff members at EPA (see App. A) indicated that emission factors for FBC are not available. The standard reference for emission factors (U.S. Environmental Protection Agency, 1984) has no information on FBC, and there is only one SO_2 AFBC emission factor in the National Emissions Data System (NEDS). The Hazardous and Trace Element Emissions System (HATREMS) has no information on trace element emissions from FBC.

A literature review was initiated by doing a keyword search on the computerized DOE/RECON information system, which accesses about 40 bibliographic and nonbibliographic data bases. Over 134 references were identified related to FBC and atmospheric emissions. Most of these references were eliminated from further consideration because their abstracts indicated that they did not contain data from which emission factors could be calculated. In addition, standard environmental bibliographies and air pollution journals were consulted to identify additional information. Information useful for trace metals comes primarily from three boilers (i.e., the B&W/Alliance AFBC boiler, the Georgetown University AFBC boiler, and the Exxon miniplant PFBC boiler) and deals only with the combustion of bituminous coal.

2.1 CONVENTIONAL POLLUTANTS

Although they are not the focus of this study, both AFBC and PFBC can apparently meet the utility boiler New Source Performance Standards (NSPS). For bituminous coal, the standard for new electric utility steam-generating units larger than 250×10^6 Btu/hr requires that (40 CFR 60, Subpart Da):*

- Particulate emissions not exceed $0.03 \text{ lb}/10^6 \text{ Btu}$, with a 99% reduction of uncontrolled emissions.
- SO_2 emissions not to exceed $1.20 \text{ lb}/10^6 \text{ Btu}$, with a 90% reduction of uncontrolled emissions, or $0.60 \text{ lb}/10^6 \text{ Btu}$, with a 70% reduction of uncontrolled emissions.
- NO_x emissions not to exceed $0.60 \text{ lb}/10^6 \text{ Btu}$ (30-day rolling average).

At the Georgetown AFBC, Fennelly et al. (1983) report an average particulate loading of $0.005 \text{ lb}/10^6 \text{ Btu}$, almost an order of magnitude lower than the NSPS limit. This unit is controlled by a baghouse, and the tests indicate collection efficiencies of 99.945-99.999%, which are well above the 99% efficiency required. However, as noted by Bubenick et al. (1981), emissions from FBC frequently contain more fine particulates

*Standards of Performance for New Stationary Sources, Standards of Performance for Electric Utility Steam Generating Units for Which Construction Is Commenced after September 18, 1978.

than emissions from conventional boilers. Thus, if EPA promulgates a National Ambient Air Quality Standard (NAAQS) for particulate matter less than $10\ \mu\text{m}$, FBC units may be greater relative contributors to atmospheric loading than conventional combustors. Although this same boiler only achieved 85-95% SO_2 removal, no special procedures were followed to achieve consistently high SO_2 removal efficiencies. The SO_2 removal efficiency is highly dependent on the ratio of the calcium in the limestone to the sulfur in the coal (Ca:S ratio). At ratios higher than five, 90% SO_2 removal could probably be achieved consistently. The operational problem is to achieve 90% SO_2 removal at a lower Ca:S ratio so as to minimize both operating costs and the volume of solid waste (Fennelly, 1984).

FBC boilers have an advantage over conventional boilers with regard to NO_x emissions. They operate at a temperature below the one at which molecular nitrogen in the coal oxidizes; conventional boilers operate above that temperature. Thus, NO_x emissions control is almost entirely a question of regulating the excess air in the combustor to prevent formation of NO_x from atmospheric nitrogen. The Georgetown AFBC routinely achieved emissions of about $0.5\ \text{lb}/10^6\ \text{Btu}$, a level below the NSPS level. Newer designs have achieved even lower levels. For example, on the basis of tests at the B&W/Alliance boiler, modification of existing designs should enable a limit of $0.2\ \text{lb}/10^6\ \text{Btu}$ to be met.

Although no tests definitely indicate that the PFBC boiler could achieve 99% particulate control, Kindya et al. (1981a) present data that show particulate collection efficiencies in the 98.7% to 99.6+% range (see App. B for estimates of PFBC emissions factors) with only cyclones being used for control. A baghouse should enable control levels well in excess of 99% to be achieved consistently.

The SO_2 emissions at the Exxon miniplant were $0.09\ \text{lb}/10^6\ \text{Btu}$ (Murthy et al., 1979). The coal used contained 1.7% sulfur and had a heating value of $13,500\ \text{Btu}/\text{lb}$, which corresponds to an uncontrolled SO_2 emission rate of about $2.5\ \text{lb}/10^6\ \text{Btu}$. Control to $0.25\ \text{lb}/10^6\ \text{Btu}$ would be required to meet NSPS. The observed emission rate meets this limit. If these results are truly indicative of full-scale operating practice, PFBC may have an advantage over AFBC in terms of the control of SO_2 emissions.

NO_x emissions at the Exxon PFBC were only $0.18\ \text{lb}/10^6\ \text{Btu}$, which is well below the NSPS limit. Here again, indications are that PFBC may have an advantage over AFBC in NO_x control. However, these seeming advantages would need to be confirmed by additional tests on other units.

2.2 TRACE ELEMENTS

The control of trace metal emissions is essentially good particulate control. Trace metals from FBC tend to be controlled to about the same degree as overall particulates (Fennelly et al., 1980). Thus, it is expected that over 99% of the trace metals could be removed if baghouses were employed. It must be emphasized, however, that this expectation needs to be confirmed by actual field measurements on full-sized FBC units, especially because arsenic, cadmium, and chromium tend to concentrate on the fine particles. These particles are more likely to penetrate a collector, even a baghouse (Mead et al., 1986).

There are no national limits for trace metal emissions for combustion units. However, EPA's multipollutant assessment of boilers may lead to promulgation of such regulations.

Table 1 presents selected trace element emission factors for AFBC and compares them with the corresponding factors for the combustion of pulverized bituminous coal in wet- and dry-bottom utility boilers. The tabulated factors were calculated from the data on the concentration of stack gases given in the references (see App. B). The assumptions made in converting the concentration data to a heat input basis are also given in that appendix. The range presented for the AFBC emission factors reflects two different assumptions regarding the volume of flue gas generated per unit of heat input. The emission factors for conventional combustion are those recommended for use in Mead et al. (1986).

TABLE 1 Approximate Trace Element Emission Rates for AFBC (10^{-4} lb/ 10^6 Btu)

Element	AFBC ^a		Conventional Utility Boiler Burning Pulverized Coal ^c	
	Emission Factor	Comparison with Conventional Boiler ^b	Dry Bottom	Wet Bottom
Antimony	0.28-34	NA	-	-
Arsenic	0.0028-0.0034	<	6.8	13.4
Beryllium	0.58-71	≈	0.81	0.81
Cadmium	0.14-0.17	<	0.44	0.45-0.70
Chromium	14-18	≈	12.5-15.7	10.2-15.7
Cobalt	4.9-6.0	NA	-	-
Copper	9.4-11.6	≈	8.5	5.7-8.5
Lead	5.7-7.1	≈	5.3	5.3
Manganese	69-86	>	30	8-30
Mercury	0.17-0.21	≈	0.16	0.16
Nickel	24-30	>	10-13	10-13
Vanadium	19-24	NA	-	-
Zinc	11-13	NA	-	-

^aSee App. B for technical information and references.

^b > = FBC emissions more than about 1.6 times greater than conventional emissions.

≈ = FBC and conventional emissions equal to within a factor of about 1.6.

< = FBC emissions less than about six-tenths of conventional emissions.

NA = not available.

^cRecommended factors are from Mead et al., 1986.

With the exception of manganese and nickel, AFBC emissions are less than or about equal to emissions from conventional utility boilers that fire pulverized bituminous coal. AFBC shows lower arsenic and cadmium emissions than convention boilers and about the same beryllium, chromium, copper, lead, and mercury emissions. In making these comparisons, the emissions are called "about equal" if the emission factor ranges overlap when multiplied by a factor in the range of 0.6 ($1/1.6 = 0.625 \approx 0.6$) to 1.6. This factor of 1.6 was chosen on the basis of data presented in Kindya et al. (1981b), who report the results of two test runs. The factor 1.6 is the average factor by which emission factors differ between two runs; therefore, it reflects the precision of the AFBC measurements and emission factors.

Nickel and manganese emissions from AFBC appear to exceed those from conventional boilers. EPA is considering nickel for regulation as a hazardous air pollutant. Whether the potential for higher nickel emissions would affect the use of AFBC would depend on the emission limits promulgated by EPA. No comparisons could be performed for antimony, cobalt, vanadium, and zinc because emission factors for conventional boilers were not available.

Although total AFBC emissions exceed the total conventional boiler emissions for the nine trace elements for which both AFBC and conventional emission factors are available, it cannot necessarily be said that AFBC presents a greater health risk. Such a conclusion would require additional information on the relative toxicity of the various chemical species in which each trace element is emitted and the emission conditions (e.g., stack heights, temperatures, and volumes). Such data speciation is not readily available, even for conventional combustors.

Table 2 presents selected trace element emission factors for PFBC and compares them with the corresponding factors for conventional combustion. The same general comments apply to Tables 1 and Table 2; details of the calculations and assumptions are provided in App. B. The emissions measurements for PFBC were taken downstream from particulate control devices. The range presented for PFBC emission factors reflects two assumptions regarding the overall collection efficiency of these control devices. Only a single estimate, rather than two as was true for AFBC, could be made of the volume of flue gas generated per unit of heat input for PFBC.

PFBC trace element emissions are always about equal to or greater than the corresponding emissions from conventional boilers. Since results from multiple tests are not presented in the references, the factor of 1.6 used for AFBC was also used to determine how PFBC emissions compared with those from conventional boilers. PFBC emissions of chromium, mercury, and nickel exceeded conventional boiler emissions. However, as was the case with AFBC, further analysis would be needed to determine whether the higher emissions are likely to result in a higher health risk.

PFBC emissions of chromium exceed those of conventional boilers by factors possibly in excess of 100. Although relatively high, it is not clear how a chromium NESHAP might affect the use of PFBC boilers until specific emission limits are proposed by EPA.

TABLE 2 Approximate Trace Element Emission Rates for PFBC (10^{-4} lb/ 10^6 Btu)

Element	Emission Factor	PFBC ^a	Conventional Utility Boiler Burning Pulverized Coal ^c	
		Comparison with Conventional Boiler ^b	Dry Bottom	Wet Bottom
Antimony	0.11-37	NA	-	-
Arsenic	2.3-7.8	≈	6.8	13.4
Beryllium	0.33-1.11	≈	0.81	0.81
Cadmium	0.78-2.6	≈	0.44	0.45-0.70
Chromium	371-1236	>	12.5-15.7	10.2-15.7
Cobalt	3.6-12.0	NA	-	-
Copper	0.4-15	≈	8.5	5.7-8.5
Lead	4.4-15	≈	5.3	5.3
Manganese	33-111	≈	30	8-30
Mercury	1.6-5.4	>	0.16	0.16
Nickel	224-747	>	10-13	10-13
Vanadium	17-57	NA	-	-
Zinc	550-1800	NA	-	-

^aSee App. B for technical information and references.

^b > = FBC emissions more than about 1.6 times greater than conventional emissions.

≈ = FBC and conventional emissions equal to within a factor of about 1.6.

NA = not available.

^cRecommended factors from Mead et al., 1986.

3 SUMMARY

Comparing Tables 1 and 2 indicates that AFBC emissions of arsenic, cadmium, chromium, mercury, nickel, and zinc would probably be lower than the corresponding PFBC emissions. For all other tabulated trace elements, the emissions are about the same for both technologies. Emissions were called "about the same" if there was any overlap in the emission factor ranges given in the two tables. Overall, AFBC seems to present a lower potential for emission of trace elements than PFBC. However, as noted previously, PFBC seems to have lower SO_2 and NO_x emissions. Whether one of the technologies offers a greater advantage from the perspective of atmospheric emissions will probably depend on which emissions are of concern.

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APPENDIX A

PERSONS CONTACTED

D. Patrick	EPA, Strategies and Air Standards Division
W. Stevenson	EPA, Emissions Standards and Engineering Division
D. Sherman	National Resources Defense Council
C. Mann	EPA, Monitoring and Data Analysis Division
T. Lahre	EPA, Strategies and Air Standards Division
B. Henschel	EPA, Industrial Environmental Research Laboratory
F. Porter	EPA, Emission Standards and Engineering Division

APPENDIX B

CALCULATIONS AND ASSUMPTIONS

ESTIMATION OF AFBC EMISSION FACTORS

Kindya et al. (1981a) report on a trace element analysis of flue gas streams from the B&W/Alliance AFBC. Their results are presented in units of $\mu\text{g}/\text{m}^3$, a measure of concentration in the flue gas, rather than in units of $\text{lb}/10^6 \text{ Btu}$, a measure of mass per unit of heat input. However, an approximate conversion can be made based on information presented in the paper.

Fly ash concentrations during sampling were about $35 \text{ g}/\text{Nm}^3$, and emissions could meet an emission limit of $0.03 \text{ lb}/10^6 \text{ Btu}$ with 99.9% control of particulates. Assuming that this percentage of control would meet this emission limit exactly, then:

$$35 \text{ g}/\text{m}^3 \times (1 - 99.9/100) \times \text{CF} = 0.03 \text{ lb}/10^6 \text{ Btu}$$

$$\text{CF} = 0.00857 \quad (\text{B.1})$$

where CF is a conversion factor. In other words, to convert from $(\mu\text{g}/\text{m}^3)$ to $(10^{-4} \text{ lb}/10^6 \text{ Btu})$, multiply by 0.00857. This factor permits approximate conversion of emissions on a concentration basis to emissions on a heat input basis.

Kindya et al. (1981a) express their results in terms of a "discharge severity" (DS), which is defined as the measured elemental concentration in flue gas divided by air DMEGs (discharge multimedia environmental goals). Since the air DMEGs are given, the emission factor (EF) for a particular element can be approximated by:

$$\text{EF} (10^{-4} \text{ lb}/10^6 \text{ Btu}) = 0.00857 \times \text{DS} \times \text{DMEG} (\mu\text{g}/\text{m}^3) \quad (\text{B.2})$$

In the preceding derivation, the assumption embodied in Eq. B.1 may be imprecise; a control efficiency slightly different from 99.9% may be needed to meet the $0.03 \text{ lb}/10^6 \text{ Btu}$ emission limit exactly. Babcock & Wilcox Co. (1985) prepared conceptual designs for AFBC utility units. The results for two designs firing eastern coals are: $1,338,816 \text{ scfm}^*$ at $5628.3 \times 10^6 \text{ Btu/hr}$ for one unit and $1,410,142 \text{ scfm}$ at $5743.3 \times 10^6 \text{ Btu/hr}$ for the second.

Using these data, an average conversion factor corresponding to CF can be calculated to be 0.00910. This value is within about 6% of the value calculated in Eq. B.1. The essential equivalence of these results indicates that the conversion factor CF is reasonably reliable.

Another estimate of the conversion factor CF can be made using the data in Kindya et al. (1981a). In their experiment, the cyclones and baghouse were used for emissions control, with the total weight of the catch for four runs given along with the

*Standard cubic feet per minute.

coal feed rates and coal heating value. Assuming that essentially all the emissions are caught, which is a reasonable assumption for a baghouse:

$$\begin{aligned}
 CF &= [(total\ catch)/(total\ heat\ input)]/[stack\ gas\ concentration] \\
 &= [589\ lb/hr/24.23\ 10^6\ Btu/hr \times 10^4\ (10^{-4}\ lb/lb)]/ \\
 &\quad [35\ g/m^3 \times 10^6\ \mu g/g] \\
 &= 0.00694 \qquad (B.3)
 \end{aligned}$$

In other words, to convert from $(\mu g/m^3)$ to $(10^{-4}\ lb/10^6\ Btu)$, multiply by 0.00694. This value of CF was used in place of the factor 0.00857 in Eq. B.2 to make a second estimate of the AFBC emission factors.

ESTIMATION OF PFBC EMISSION FACTORS

Murthy et al. (1978, 1979), and Kindya et al. (1981b) provide emissions data for the Exxon miniplant PFBC boiler. After two conventional cyclones, the concentration of particulates was 1.2 gr/scf or $1.9\ lb/10^6\ Btu$. Using this equivalence, a conversion factor, CF', for converting tabulated trace element concentrations from $\mu g/m^3$ to $10^{-4}\ lb/10^6\ Btu$ can be calculated:

$$\begin{aligned}
 CF' &= \frac{1.9}{1.2} \left(\frac{lb/10^6\ Btu}{gr/ft^3} \right) \times \frac{1}{0.0648} \left(\frac{gr}{g} \right) \times 0.0283 \left(\frac{ft^3}{m^3} \right) \\
 &\quad \times 10^{-6} \left(\frac{g}{\mu g} \right) \times 10^4 \left(\frac{10^{-4}\ lb}{lb} \right) \\
 &= 0.00692 \qquad (B.4)
 \end{aligned}$$

In other words, to convert from $(\mu g/m^3)$ to $(10^{-4}\ lb/10^6\ Btu)$, multiply by 0.00692.

Kindya et al. (1981b) give the most recent and complete data, the flue gas concentrations being taken after the third in a series of cyclones. The first cyclone is part of the PFBC process; its catch is recycled to the combustor so that the potential uncontrolled atmospheric emissions would occur after this cyclone. There is another emission vent from the regenerator to the atmosphere, but this waste stream "does not represent a true emission stream." Hence, it was ignored in this analysis. To calculate uncontrolled emissions from the PFBC unit, the overall efficiency of the second and third cyclones is required. Kindya et al. (1981b) provide data for this determination.

Particulate loadings at the inlet of the second cyclone are from 8 to $12\ g/Nm^3$. Loadings at the outlet of the third cyclone are from 0.03 to $0.15\ g/Nm^3$. Assuming that

the maximum and minimum of the first range correspond to the maximum and minimum of the second, a reasonable range for the throughput, ϵ , and efficiency, η , of the second and third cyclones can be calculated:

$$0.15/12 = 0.0125 \leq \epsilon \leq 0.00375 = 0.03/8 \quad (\text{B.5})$$

which corresponds to:

$$98.75\% \leq \eta \leq 99.63\% \quad (\text{B.6})$$

Using these estimates of control efficiency and the conversion factor CF' , a range of emissions factors corresponding to the two values of ϵ can be calculated:

$$EF (10^{-4} \text{ lb}/10^6 \text{ Btu}) = 0.00692 \times [DS \times DMEG (\mu\text{g}/\text{m}^3)]/\epsilon \quad (\text{B.7})$$

This equation is similar to Eq. B.2, since the data are presented in terms of discharge severity DS.

APPENDIX B REFERENCES

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